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(54) Stable aqueous suspension concentrate composition.

(57) Stable aqueous flowable compositions, comprising, about 10.0% to 60.0%, pesticide; about 1.0%, to 10.0% sodium or calcium lignin sulfonate; about 0.05% to 0.5% thickening agent; about 0.05% to 1.0% thixotropic agent; about 0.1% to 1.0% antiseptic agent; and sufficient water to total the composition to 100%

The compositions are particularly effective for preparing stable aqueous suspension concentrates of pesticides with melting points less than 60°C, specifically pendimethalin.

Thixotropic agents suitable for use in the compositions of the present invention include clays and silica derivatives, with sodium montmorillonite being preferred for suspension concentrates of pendimethalin.

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STABLE AQUEOUS SUSPENSION CONCENTRATE COMPOSITIONSBACKGROUND OF THE INVENTION

Suspension concentrate pesticidal compositions or aqueous flowable pesticidal compositions are concentrated suspensions of water-insoluble pesticides and mixtures of pesticides in an aqueous system. The present invention relates to stable such aqueous suspension concentrate compositions.

These aqueous compositions frequently contain about 10% to 80%, by weight, of a solid pesticide or mixture of solid pesticides, thereby providing a method for handling those pesticides which are relatively water-insoluble in an aqueous medium. Since these types of compositions have the desirable characteristics of a thick liquid, they may be poured or pumped. Thus, some of the problems, like dusting that is possible in solid compositions of wettable powders and granulars, are avoided. Further, these aqueous-based concentrates also have the added advantage of not requiring the use of organic solvents, often present in emulsifiable concentrates.

For these reasons, it is desirable to formulate pesticides into suspension concentrates or aqueous flowables. However, such formulations have their own problems such as gelling, caking and settling, as well as problems because of the physical and chemical characteristics of the pesticide or mixture of pesticides. For instance, the dinitroaniline, pendimethalin, is somewhat difficult to formulate and several references have tried to address these formulation problems.

The problems associated with the development of suspension concentrate compositions containing low-melting active ingredients, alone or in combination with higher melting active ingredients, are described in German Patent Application DE 3302648 A1. German Patent Application DE3302648 A1 tries to deal with the problems of an aqueous mixed dispersion of a low-melting active ingredient in a solvent of phthalic acid C₁-C₁₂ alkyl esters in combination with an aqueous suspension concentrate containing one or more active ingredients as an alternative to a suspension concentrate containing low-melting active ingredients, such as pendimethalin [N-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine]. The reason for the alternative approach of that application is the inability to prepare stable suspension concentrates by various techniques, including those of European Patent Application 0 33291 2. That EPO application describes insecticidal suspension concentrate compositions of phosalone and adjuvants which may be prepared with molten insecticide.

Pendimethalin is known to exist at ambient temperatures in the form of two distinct polymorphs, a yellow microcrystalline form and an orange macrocrystalline form. United States Patents 4,082,537 and 4,150,969 describe stable wettable powder compositions of pendimethalin in which pendimethalin is stabilized in the yellow crystal form, to avoid the problems encountered when the larger orange crystal form is present in such wettable powder compositions. However, attempts to prepare aqueous suspension concentrate compositions of stabilized pendimethalin by utilizing preparation methods acceptable for wettable powder compositions results in unstable aqueous suspension concentrate compositions which tend to grow larger crystals upon aging.

The present invention successfully formulates pendimethalin, as well as other pesticides with melting points less than 60°C, into stable aqueous suspension concentrate compositions. The compositions of the present invention are unique in that highly purified pesticide, such as pendimethalin in the yellow crystal form, is formulated with the specialized surfactants of this invention into stable aqueous suspension concentrate compositions or aqueous flowable compositions.

It is an object of the present invention, therefore, to provide stable aqueous suspension concentrate compositions or aqueous flowable compositions of low-melting pesticides (melting point less than 60°C). Further, it is an additional object of the present invention to provide aqueous suspension concentrate compositions or aqueous flowable compositions of pendimethalin, wherein said pendimethalin is in the yellow microcrystalline form.

It is another object of the present invention to provide methods for preparing such stable aqueous suspension concentrate compositions or aqueous flowable compositions of pesticides with melting points less than 60°C, specifically pendimethalin.

SUMMARY OF THE INVENTION

The present invention relates to stable aqueous suspension concentrate compositions or aqueous flowable compositions comprising low-melting pesticides, with melting points less than 60°C, alone or in combination with pesticides having melting points higher than 70°C. Further, the compositions of the present invention include as one such low-melting pesticide pendimethalin in the yellow microcrystalline form.

Stable compositions of the present invention provide, on a weight to weight basis, improved aqueous flowable compositions, comprising, about 10.0% to 60.0%, pesticide; about 1.0%, to 10.0% sodium or calcium lignin sulfonate; about 0.05% to 0.5% thickening agent; about 0.05% to 1.0% thixotropic agent; about 0.1% to 1.0%, on a weight basis, antiseptic agent; and sufficient water to total the composition to 100%.

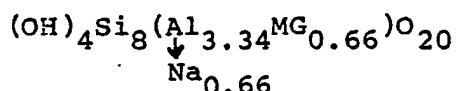
DETAILED DESCRIPTION OF THE INVENTION

The compositions of this invention are particularly effective for preparing stable aqueous suspension concentrates of pesticides having low-melting points, about 60°C or less. Further, it has been found that if the low-melting active pesticide contains impurities, which oftentimes lower the melting point, that removal of these impurities by conventional methods, such as crystallization, washing, extraction or the like, prior to the preparation of the suspension concentrate is preferable.

Sodium or calcium lignin sulfonates suitable for use in the compositions of the present invention are conventionally found for use in the lumber chemical industry. Commercially, lignin sulfonates are available in both solid and liquid form, and either may be used in the compositions of the present invention. Some lignin sulfonates contain reductive saccharides, as well as saccharide derivatives and inorganic salts. It has been found that reductive saccharides are not preferable for this invention because they sometimes allow mold to grow and lead to caking during storage. Therefore, if present, they should be removed prior to use.

Thickening agents useful for the preparation of the compositions of the present invention generally include polysaccharide gums such as Xanthan gum. Further, sodium carboxymethyl cellulose, guar gum, gum arabic, sodium alginate, polyvinyl alcohol, and the like, may also be used.

30 Thixotropic agents suitable for use in the compositions of the present invention include clays and silica derivatives, with sodium montmorillonite being preferred for suspension concentrates of pendimethalin. This compound can be extracted from a natural swelling type clay by a physical/chemical process and is a natural colloidal hydrated aluminum-silicate, having the following general formula:



40 Antiseptic agents which may be used in preventing microorganism growth in the compositions of this invention include organic iodine compounds, benzoic acids, esters and salts and sodium dehydroacetate. The following examples further illustrate the present invention but are not limitative thereof.

43 EXAMPLES I-22

Preparation of aqueous suspension concentrate compositions containing pendimethalin

50 Pendimethalin, I30.5 g (96% purity), II.0 g of the various surfactants listed in Table I, 0.55 g of sodium montmorillonite and I2I g of water are premixed, using an Osterizer (homogenizer), for five minutes at 15 ± 50°C. The resulting mixture is sand-ground, using a water-jacketed sand-grinder, for one to two hours. Antiseptic agents listed in Table II and 0.16 g of thickening agents listed in Table III are dissolved in 10 g of water, and the solution is then admixed with the ground mixture prepared above. The resulting aqueous suspension concentrate composition is then packaged.

Utilizing the above procedure and the surfactants, antiseptic agents and thickening agents listed in Tables I, II and III, yields the aqueous suspension concentrate compositions listed in Table IV.

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TABLE I
Surfactants

	<u>Designation</u>	<u>Description</u>
15		P.O.E. ¹ styryl phenol ether
	A-1	Dialkyl sulfosuccinate
		P.O.E. alkyl arlether sulfate
20		P.O.E. stryl phenol ether
	A-2	Dialkyl sulfosuccinate
		P.O.E. alkyl arylether sulfate
25		P.O.E. stryl phenyl ether polymer
	A-3	Dialkyl sulfosuccinate
	A-4	Polyoxyalkylene glycol sulfate
		P.O.E. stryl phenyl ether
30	A-5	P.O.E. alkyl aryl ether
	A-6	Fatty acid polymer
	A-7	P.O.E. stryl phenyl ether sulfate
35	A-8	Nonionic surfactant
	A-9	Nonionic surfactant
	A-10	Naphthalene sulfonate
	A-11	Lauryl sulfate
40	A-12	Dodecyl benzene sulfonate
	A-13	P.O.E. octyl phenyl ether
	A-14	Lignin sulfonate-Ca (61%)
45	A-15	Lignin sulfonate-Na (91%)

¹Polyoxyethylene

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15 TABLE II
Antiseptic agents

<u>Designation</u>	<u>Description</u>	<u>Composition rate</u>
B-1	Organic iodine compound	0.2%
B-2	Sodium dehydroacetate	0.2%
B-3	Sodium benzoic acid ester	0.2%

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35 TABLE III
Thickening agent

<u>Designation</u>	<u>Description</u>
C-1	Xanthan gum, polysaccharide gum
C-2	CMC-Na, Sodium carboxymethyl cellulose
C-3	Arabic gum, Arabin
C-4	PVA, Polyvinyl alcohol
C-5	Guar gum, Cyamoposis gum
C-6	Alginate-Na, Sodium alginate

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TABLE IV
Formulation

10	No	Antiseptic agent	Surfactants	Thickening agents
15	1	B-1	A-1	C-1
	2		A-2	
20	3		A-3	
	4		A-4	
25	5		A-5	
	6		A-6	
30	7		A-7	
	8		A-8	
	9		A-9	
35	10		A-10	
	11		A-11	
	12		A-12	
40	13		A-13	
	14		A-14	
	15		A-15	
45	16	B-2	A-15	
	17	B-3		
	18	B-1		C-2
	19			C-3
	20			C-4
	21			C-5
	22			C-6

45 EXAMPLE 23

Stability of aqueous suspension concentrate compositions

50 The compositions prepared in Examples I-22 are maintained at 45°C in an oven for one week. Observation is made with respect to particle growth and caking, according to the methods described below:

Particle growth

55 Aged samples are observed microscopically and rated according to the rating system described hereinbelow. When crystals are found, length of crystal is measured.

Caking

Aged samples in glass bottle are inverted ten times and checked if caking is still evident in the bottom of the bottle after the procedure. Then, the same rating system provided hereinbelow is used to rate caking.

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Processing characteristics

Difficulties encountered during preparation of each sample are noted and recorded.

10 The results of these experiments, summarized in Table V, demonstrate improved stability of the compositions of the present invention.

Rating System + + + Very significant

15 + + Significant

+ Visible difference

± Slight

- None

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TABLE V

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	Composition of example no	Particle growth	Caking	Difficulty in grinding
10	1	+++	++	D with foam
	2	+++	++	D with foam
	3	+++	++	D with foam
15	4	+++	++	D with foam
	5	+++	++	D with foam
20	Comparative Compositions	6	+++	D with foam
	7	+++	++	D with foam
25		8	++	+++ D with foam
		9	+++	++ D with foam
		10	++	+++ D with sticky substance
30		11	++	+++ D with sticky substance
		12	++	+++ D with sticky substance
		13	+++	++ Easy
35		14	+	+
		15	+	- Easy
		16	+	- Easy
40	Compositions of the Invention	17	+	- Easy
		18	+	+
		19	+	- Easy
		20	+	- Easy
45		21	+	++ Easy
		22	+	- Easy

50 Claims

I. An aqueous flowable pesticidal concentrate composition comprising, on a weight to weight basis: about 10.0% to 60.0% pesticide; about 1.0% to 10.0% sodium or calcium lignin sulfonate; about 0.05% to 0.5% thickening agent; about 0.1% to 1.0% antiseptic agent; about 0.05% to 1.0% montmorillonite; and the remainder water to total said composition to 100%.

55 2. A composition according to Claim I, wherein said pesticide has a melting point less than 60°C.

3. A composition according to Claim 2, wherein said pesticide is N-(*i*-ethylpropyl)-2,6-dinitro-3,4-xylidine, pendimethalin, in the yellow microcrystalline form.
4. A composition according to Claim 3, wherein said pendimethalin has been purified to remove impurities, which lower its melting point, by crystallization, washing, extraction or mixtures thereof.
5. A composition according to Claim 4 wherein said thickening agent is Xanthan gum.
6. A composition according to Claim 4, wherein said thickening agent is selected from the group consisting of sodium carboxymethyl cellulose, gum arabic, guar gum, sodium alginate or polyvinyl alcohol.

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EUROPEAN SEARCH REPORT

EP 87 10 6430

DOCUMENTS CONSIDERED TO BE RELEVANT									
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)						
X,D	DE-A-3 302 648 (HOECHST) * Page 7, line 21 - page 8, line 7; page 15, examples 6,9; page 16, lines 13-14 *	1-6	A 01 N 25/04 A 01 N 33/18						
X	US-A-3 157 486 (T.S. HARRISON) * Claims *	1-6							
X	EP-A-0 110 174 (HOECHST) * Claims; examples *	1-6							
A,D	US-A-4 082 537 (J.J. DUDKOWSKI) * Column 2, lines 6-31 *	3							

TECHNICAL FIELDS SEARCHED (Int. Cl.4)									
A 01 N									
<p>The present search report has been drawn up for all claims</p> <table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 33%;">Place of search</td> <td style="width: 33%;">Date of completion of the search</td> <td style="width: 34%;">Examiner</td> </tr> <tr> <td>THE HAGUE</td> <td>28-09-1987</td> <td>DECORTE D.</td> </tr> </table>				Place of search	Date of completion of the search	Examiner	THE HAGUE	28-09-1987	DECORTE D.
Place of search	Date of completion of the search	Examiner							
THE HAGUE	28-09-1987	DECORTE D.							
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document							